

# Comment on: “Luminescence spectra of quantum dots in microcavities. ”

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(Dated: May 31, 2010)

In this comment we show that there is a direct connection between coherent exchange of energy among light and matter and the emission spectrum of a microcavity quantum dot system as modeled in Phys. Rev. B **79**, 235325 (2009) by F. P. Laussy, E. del Valle, and C. Tejedor. To do so, we show that in their model the necessary and sufficient conditions for having eigenvalues with non-zero imaginary parts in the propagator of the bare mode populations, are the same as for having strong coupling in the emission spectrum. This amounts to saying that, whenever there is strong coupling there will be oscillating frequencies in the dynamics of the populations. These conditions are valid both for the case where matter is treated as bosonic or fermionic, in the spontaneous emission case.

PACS numbers: 42.50.Ct, 78.67.Hc, 42.55.Sa, 32.70.Jz

Laussy and coworkers<sup>1-3</sup> studied the effects of considering incoherent pumping of both excitons and photons in a microcavity quantum dot system. In particular, they clearly show the nontrivial effects of such pumping mechanisms in the emission spectrum of the system. The system is modeled using the following quantum master equation (in units where  $\hbar = 1$ ):

$$\begin{aligned} \frac{d}{dt}\rho = & i[\rho, H] + \sum_{c=a,b} \frac{\gamma_c}{2} (2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c) \\ & + \sum_{c=a,b} \frac{P_c}{2} (2c^\dagger \rho c - cc^\dagger \rho - \rho cc^\dagger), \end{aligned} \quad (1)$$

where  $H = \sum_{c=a,b} \omega_c c^\dagger c + g(a^\dagger b + ab^\dagger)$ .  $a, a^\dagger$  are photonic boson operators and  $b, b^\dagger$  can be either bosonic or fermionic matter operators. The quantities  $\omega_c$  are the bare energies of the light and matter,  $g$  is the coupling constant between them,  $\gamma_c$  are the decaying rates of the cavity and the emitter and  $P_c$  are the rates at which they are being incoherently pumped.

For the case where the exciton is modeled as a boson Laussy *et. al.* define a criteria for having strong coupling (SC)<sup>1</sup>:

$$g > |\Gamma_-|, \quad (2)$$

where  $4\Gamma_\pm = \Gamma_a \pm \Gamma_b = (\gamma_a - P_a) \pm (\gamma_b - P_b)$ . We will show, that the same criterion of the above equation, is a necessary and sufficient condition for having oscillatory frequencies in the propagator of the bare mode populations. For convenience we re-write equation (12) of reference [1] as follows:

$$\begin{aligned} \frac{d}{dt} \begin{pmatrix} n_a \\ n_b \\ \beta \\ \alpha \end{pmatrix} = & \begin{pmatrix} P_a \\ P_b \\ 0 \\ 0 \end{pmatrix} + \\ & \begin{pmatrix} -\Gamma_a & 0 & -2g & 0 \\ 0 & -\Gamma_b & 2g & 0 \\ g & -g & -2\Gamma_+ & -\Delta \\ 0 & 0 & \Delta & -2\Gamma_+ \end{pmatrix} \times \begin{pmatrix} n_a \\ n_b \\ \beta \\ \alpha \end{pmatrix}, \end{aligned} \quad (3)$$

where  $n_a = \langle a^\dagger a \rangle$ ,  $n_b = \langle b^\dagger b \rangle$ ,  $\alpha = \Re(\langle a^\dagger b \rangle)$ ,  $\beta = \Im(\langle a^\dagger b \rangle)$ , and  $\Delta = \omega_a - \omega_b$ . The above equation can be written in a more compact form as:  $\frac{d}{dt}\mathbf{w}(t) = \mathbf{f} + \mathbf{A}\mathbf{w}(t)$ , where  $\mathbf{A}$  corresponds to the coefficient matrix,  $\mathbf{w}(t)$  is the vector which contains the single time mean values of interest, *i.e.*,  $\mathbf{w}(t) = (n_a(t), n_b(t), \beta(t), \alpha(t))^T$  and  $\mathbf{f} = (P_a, P_b, 0, 0)^T$ . The formal solution of this equation is:

$$\mathbf{w}(t) = e^{\mathbf{A}t}\mathbf{w}(0) + \mathbf{A}^{-1}(e^{\mathbf{A}t} - 1)\mathbf{f}, \quad (4)$$

where  $e^{\mathbf{A}t}$  is the propagator of the last equation. The eigenvalues of the matrix  $\mathbf{A}$  are given by:

$$\begin{aligned} \lambda(\mathbf{A})_{\pm, \pm} = & -2\Gamma_+ \pm \sqrt{2}\sqrt{a \pm \sqrt{b}}; \\ a = & \Gamma_-^2 - g^2 - \frac{\Delta^2}{4}; \\ b = & (\Gamma_-^2 - g^2)^2 + (g^2 + \Gamma_-^2)\frac{\Delta^2}{2} + \frac{\Delta^4}{16}. \end{aligned} \quad (5)$$

In resonance condition ( $\Delta = 0$ ) they simplify to:

$$\lambda(\mathbf{A}) = \{-2\Gamma_+, -2\Gamma_+, -2\Gamma_+ + 2iR_0, -2\Gamma_+ - 2iR_0\}, \quad (6)$$

where  $R_0 = \sqrt{g^2 - \Gamma_-^2}$ . Notice that if  $R_0$  is a positive number, the condition (2) is automatically fulfilled, *i.e.*, light and matter are in the SC regime. Then, if  $R_0 > 0$ , there will be imaginary frequencies in the propagator that will lead to oscillations in the populations. As a consequence of choosing as variables the real and imaginary parts of the coherence  $\langle a^\dagger b \rangle$ , the propagator in resonance condition is given by a block diagonal matrix, with form:

$$e^{\mathbf{A}t} = \begin{pmatrix} \mathbf{M}_{3 \times 3}(t) & \mathbf{0} \\ \mathbf{0} & e^{-2\Gamma_+ t} \end{pmatrix}. \quad (7)$$

Notice that the variable  $\alpha$  decouples from equation (3) and undergoes exponential decay  $\alpha(t) = e^{-2\Gamma_+ t}\alpha(0)$ . The square matrix  $\mathbf{M}_{3 \times 3}(t)$  is given by:

$$\mathbf{M}_{3 \times 3}(t) = e^{-2\Gamma t} \times \begin{pmatrix} \frac{(R_0 \cos(R_0 t) - \Gamma_- \sin(R_0 t))^2}{R_0^2} & \frac{g^2 \sin^2(R_0 t)}{4R_0^2} & \frac{2g \sin(R_0 t)(\Gamma_- \sin(R_0 t) - R_0 \cos(R_0 t))}{R_0^2} \\ \frac{g^2 \sin^2(R_0 t)}{4R_0^2} & \frac{(\Gamma_- \sin(R_0 t) + R_0 \cos(R_0 t))^2}{R_0^2} & \frac{2g \sin(R_0 t)(\Gamma_- \sin(R_0 t) + R_0 \cos(R_0 t))}{R_0^2} \\ \frac{g \sin(R_0 t)(R_0 \cos(R_0 t) - \Gamma_- \sin(R_0 t))}{R_0^2} & -\frac{g \sin(R_0 t)(\Gamma_- \sin(R_0 t) + R_0 \cos(R_0 t))}{R_0^2} & \frac{(g^2 \cos(2R_0 t) - \Gamma_-^2)}{R_0^2} \end{pmatrix}. \quad (8)$$

The above equation explicitly shows that in SC the populations undergo oscillations with frequencies proportional to  $2R_0$ .

In this paragraph, we re-examine the results in section V of ref. [1]. In figure 6 of the same article they present the dynamics of the average photon number  $n_a$  for different initial conditions. In particular, they claim that in strong coupling there is no oscillation in such observable for the initial condition of one upper polariton with the parameters  $\Delta = P_a = P_b = 0$ ,  $\gamma_a = 1.9g$  and  $\gamma_b = 0.1g$ . When one plugs their parameters into the propagator, equation (8), one sees that there are oscillating frequencies, yet the dynamics seems to be simply decaying in the populations  $n_a$  and  $n_b$ . When their difference is plotted it is seen that it actually oscillates showing that indeed both terms contain imaginary frequencies (see the left panel of fig. 1). One might wonder why the oscillations are not clearly visible in the populations. The reason is that the spectrum of  $\mathbf{A}$  contains two purely real eigenvalues that will hinder the other two which are complex. This is further clarified when one takes the Fourier transform of the propagator and applies it to a given initial condition. In the right panel of fig. 1 we plot the Fourier components of  $n_a$  for the parameters of the left panel of the same figure. It is clearly seen how the widened Lorentzian around zero hides the rest of the contributions at  $\pm 2R_0 \approx \pm 1.78606g$ . The reason why  $n_b - n_a$  clearly shows oscillations, with period  $T = 2\pi/(2R_0)$ , is precisely that the zero frequency component cancels out after the subtraction.

Summarizing the above discussion, we have shown that for the quantum master equation (1) the following statements are equivalent:

- The propagator of the bare mode populations has imaginary frequencies.
- The system, as modeled by equation (1), is in SC regime.

This implies that, whenever oscillations are observed in the dynamics of the bare mode populations the system will be in SC regime. In the case where one models the exciton with a fermionic operator ( $b = \sigma$ ), the spectrum of photoluminescence cannot be obtained analytically when the effects of incoherent pumping are present. When  $P_a = P_b = 0$  one can obtain exactly the eigenvalues of the dynamical matrix involved in the equations of the quantum regression theorem. del Valle *et. al.* define  $n$ th

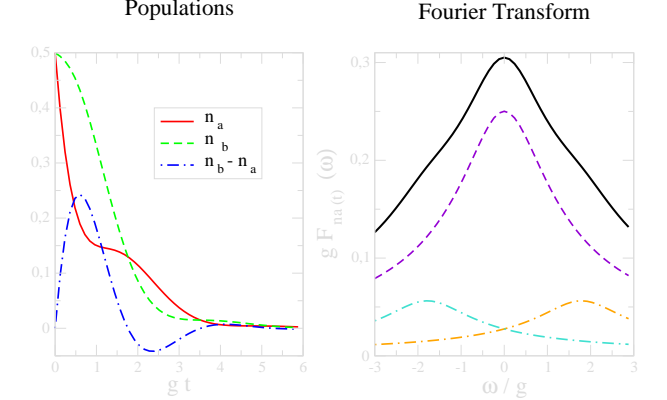


FIG. 1: (left panel) Time evolution of the populations of the photons  $n_a$ , excitons  $n_b$  and their difference for the parameters  $\Delta = P_a = P_b = 0$ ,  $\gamma_a = 1.9g$  and  $\gamma_b = 0.1g$ . The initial condition was an upper polariton,  $\langle a^\dagger b \rangle(0) = n_a(0) = n_b(0) = 1/2$ . (right panel) The dashed lines are the components of the Fourier transform of the signal  $n_a(t)$ . The solid black line is the sum of the components of the signal, *i.e.*, the Fourier transform of  $n_a(t)$ .

order SC as the following condition<sup>2</sup>:

$$g > |\Gamma_-|/\sqrt{n} \quad (9)$$

Here, we first show that the condition above is also necessary and sufficient to have imaginary frequencies, in the dynamics of the populations and coherences in the same model. To do so, we use the notation defined in [4]. We arrange the populations and coherences of the density matrix  $\rho$  as follows:  $\mathbf{u} = (\rho_{G0,G0}, \rho_{G1,G1}, \rho_{X0,X0}, \beta_1, \alpha_1, \dots, \rho_{Gn,Gn}, \rho_{Xn-1,Xn-1}, \beta_n, \alpha_n, \dots)^T$ ,  $\alpha_n = \Re(\rho_{Gn,Xn-1})$ ,  $\beta_n = \Im(\rho_{Gn,Xn-1})$  and  $\rho_{im,jn} = \langle i, m | \rho | j, n \rangle$ . Where  $i, j$  are either  $G$  (ground state) or  $X$  (exciton state) and  $n, m$  are integers representing the number of photons in the Fock state.

The equation of motion of  $\mathbf{u}$  is

$$\frac{d}{dt} \mathbf{u} = \mathbf{B} \mathbf{u}. \quad (10)$$

The structure of the matrix  $\mathbf{B}$  will consist of block diagonal terms of sizes  $1 \times 1$  (only one block, corresponding to the vacuum  $\rho_{G0,G0}$ ),  $4 \times 4$  and off diagonal terms *over* the diagonal of the matrix<sup>4</sup>. This implies that to reduce

the matrix  $\mathbf{B}$  to an upper triangular matrix it is necessary to rotate each diagonal block and the blocks above it. Once the matrix has an upper triangular form their eigenvalues are simply given by the elements of the diag-

onal. Summarizing, the eigenvalues of the whole matrix  $\mathbf{B}$  are simply the eigenvalues of the blocks  $1 \times 1$  and  $4 \times 4$ . The structure of the blocks  $4 \times 4$  corresponding to the dynamical equations of the  $n$ th excitation manifold is:

$$\mathbf{B}|_{4 \times 4} = \begin{pmatrix} -n\gamma_a & 0 & -2g\sqrt{n} & 0 \\ 0 & -((n-1)\gamma_a + \gamma_b) & 2g\sqrt{n} & 0 \\ g\sqrt{n} & -g\sqrt{n} & -\frac{\gamma_b + (2n-1)\gamma_a}{2} & -\Delta \\ 0 & 0 & \Delta & -\frac{\gamma_b + (2n-1)\gamma_a}{2} \end{pmatrix}. \quad (11)$$

The eigenvalues of the above matrix are given by:

$$\begin{aligned} \lambda(\mathbf{B}|_{4 \times 4})_{\pm, \pm} &= -\frac{\gamma_b}{2} - n\gamma_a + \frac{\gamma_a}{2} \pm \sqrt{2}\sqrt{c \pm \sqrt{d}}; \\ c &= \Gamma_-^2 - ng^2 - \frac{\Delta^2}{4}; \\ d &= g^4 n^2 + 2g^2 n \left( \frac{\Delta^2}{4} - \Gamma_-^2 \right) \\ &\quad + \left( \frac{\Delta^2}{4} + \Gamma_-^2 \right)^2. \end{aligned} \quad (12)$$

Notice that all equations derived for fermionic matter reduce to the boson matter model when  $n = 1$  and  $P_a = P_b = 0$ . However this correspondence breaks down with finite pumping or for higher excitation manifolds. In order to have  $c \pm \sqrt{d} \Big|_{\Delta=0} < 0$  it is necessary that  $g > |\gamma_a - \gamma_b|/(4\sqrt{n}) = |\Gamma_-|/\sqrt{n}$ . That is, the condition for having  $n$ th order strong coupling is the same as for having imaginary eigenvalues in the propagator of equation (10).

We would like to emphasize that the emission spectrum of the system is a robust quantity, whereas the

dynamics of the populations, in general, depends on the initial conditions. Yet, the propagators of the dynamical systems (3) and (10) are also robust features that show a similar dependence on the parameters that the emission spectrum presents.

It is important to be able to correlate the dynamical regimes (SC or weak coupling, WC) with the dynamics of the populations and coherences. For instance, the study of the entanglement between the exciton and photonic subsystems will require the knowledge of the elements of the density matrix. In the SC regime one will expect entanglement sudden death and re-birth while in WC one will see how the entanglement either vanishes in a finite time or approaches asymptotically to zero<sup>5</sup>. Finally, we would like to emphasize that the connections found between population dynamics and the emission spectrum of the system are particular to the model given by equation (1). In general, one cannot expect them to be related, and one can find models where the frequencies of the propagator and the first order correlation function depend in a different way on the system parameters<sup>6-8</sup>.

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